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ALKALI METAL FILMS TRANSPARENT IN THE SCHUMANN REGION[1]

BY W. H. WATSON[2] AND D. G. HURST[3]

Abstract

Sodium and potassium films deposited from a molecular beam on fluorite at liquid air temperature have been found to be transparent in the Schumann region. For $\lambda > 1400$ Å no transparency has been found in the case of lithium. A technique for producing these films is described. The theoretical bearing of these results is discussed.

It is well known from the experiments of R. W. Wood (12, 13), that the alkali metals when deposited as coherent films on quartz are transparent to ultra-violet light of wave-length less than the fairly well defined value characteristic of each metal, *viz.*, caesium 4400, rubidium 3600, potassium 3150, sodium 2100, lithium 2050 Å. Wood reported that all the metals, with the possible exception of caesium, are transparent to ultra-violet light of wave-length 1860 Å or greater. The experimental facts have been considered from the point of view of the theory of electrons in solids (3, 14), and in this connection it seemed to the writers desirable to extend experimental knowledge by investigating the transmission of light in the Schumann region through films of some of these metals. The present paper describes the technique developed by the writers for producing coherent films on fluorite before the slit of a vacuum spectrograph, and gives an account of the results obtained with sodium, potassium and lithium. A notice of the results with sodium and potassium has already appeared in Nature (10). So far the writers have not attempted to measure absorption coefficients.

Apparatus

The apparatus consists of three main sections: (i) the vacuum grating spectrograph (Hilger E50); (ii) the light source (the hydrogen discharge tube already described in this Journal (9)); and (iii), between (i) and (ii), separated from them by fluorite windows, the chamber in which the films are prepared. We shall call this third section the alkali-metal chamber. It is shown in Fig. 1. The upper left is a section perpendicular to the light beam; the upper right is the vertical section containing the light beam; and the lower left is the horizontal section containing the light beam. Metal is shaded

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diagonally, glass is left clear, fluorite is cross-hatched. The end of the discharge tube is shown in the upper right section above *H*.

The alkali metal is deposited on fluorite cooled to liquid air temperature. *A* is a tube to hold the liquid air, and the fluorite is held in the metal support

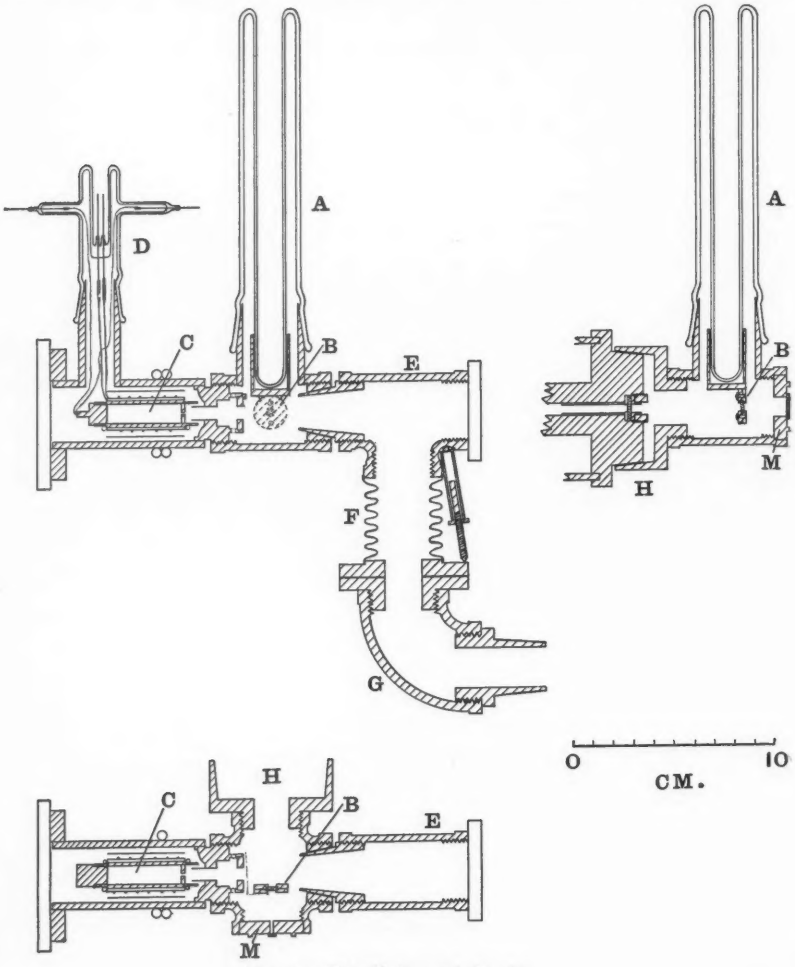

FIG. 1. *The alkali-metal chamber.*

B which slips over a copper sleeve on *A*. The copper was formed electrolytically on the silvered glass and turned in the lathe. The metal film is deposited when *A* is rotated so that the fluorite faces the oven *C* which is a steel tube supported on a light framework of Pyrex. The oven is heated by a coil of tungsten wire wound on a Pyrex tube fitting over the oven, and its temper-

ature is known by means of a thermocouple. This arrangement is quite suitable for all the alkali metals except lithium. For this metal, the oven is supported on tungsten wires and is heated by electron bombardment. The enveloping chamber, constructed wherever possible from plumbing fittings, is evacuated by its own pumping system consisting of an oil diffusion pump backed by a Hyvac. Wax seals are used to make connections except on the cone at *A* which is sealed with grease.

In order to give the freedom necessary in aligning the light source with the spectrograph, a "sylphon" *F* forms part of the pump line and the final connection is made by waxing together the brass plates on *F* and *G*, the latter being rigidly fixed. The slit of the spectrograph is enclosed when *M* presses against the end of the spectrograph and is sealed with plasticine.

The discharge tube described previously (9) has been altered in the following respects—a Pyrex tube is fitted into the exit hole to decrease sputtering, and the fluorite window is protected by a disc of glass with a hole in its centre. These are shown in Fig. 1. The writers found that great care is necessary in making and mounting the capillary of the discharge tube, otherwise fracture occurs after a short period of running.

Procedure

Sodium and potassium were triply distilled *in vacuo* and sealed into small tubes each of which contained about 0.05 gm. The alkali-metal chamber was filled with nitrogen, opened behind the oven and a stream of gas allowed to pass through. The glass tube containing the charge for the oven was broken open at both ends, placed in a helix of nichrome wire in which the metal was electrically heated, and as soon as it melted was forced by an iron plunger into the oven. The system was closed and evacuated. The oven was warmed to a suitable temperature (about 300° C. for sodium, 200° C. for potassium) found approximately by calculation to give deposition of metal at the rate of 100 atomic layers a minute. The fluorite surface at *B*, on which the film was to be deposited, was exposed to light from the hydrogen tube in order to clean it (7). The fluorite was then turned to face the oven and exposed for 10 min. at least. In order to ensure that enough metal had been deposited, the film was turned into the path of the light, and unless thicker films were desired the film was considered complete when the visible central image had been extinguished. The thinnest films were of the order of 1000 atomic layers. Unless the fluorite was cooled to liquid air temperature the writers did not succeed in making proper films. At higher temperatures the films were not coherent.

The photographs were taken on process film oiled with Nujol (4, p. 61).

Results

Examples of the spectra for sodium and potassium films have already been published (10). The whole spectrum transmitted by sodium could be photographed on one film with the proper setting of the grating. It was found that transmission began at 2100 Å and continued throughout the Schumann

region,—photographs with films of different thicknesses indicated that there is some absorption in the range 1400–1250 Å. In order to establish that the potassium films were proper ones, the practice was adopted that after the photograph of the Schumann region had been made, the grating was turned so as to show 3100 Å about the middle of the spectrum. Weak absorption by potassium films begins about 1700 Å and increases to the short waves. In the case of lithium the authors have made what they believe are good films and have not succeeded in obtaining evidence of their transparency, although weak transmission of wave-lengths less than 1400 Å might have escaped detection because of the weakness of their spectrum in this region (due to the large number of surfaces traversed by the light, cf. (6)). For the metals sodium, potassium, rubidium, caesium, the critical wave-length is proportional to the square of the lattice constant (8), and this leads one to expect no transparency in lithium for wave-lengths > 1400 Å. The transmission beginning at 2050 Å reported by Wood does not fit this law.†

The short wave ends of the transmission bands have wave-lengths roughly proportional to the *first* power of the lattice constant (a). This is shown in Table I. The choice of λ is decidedly arbitrary; nevertheless it is difficult to make the choice so as to fit the square law, because 1860 Å for caesium is probably an overestimate. It may be pointed out that the same relation holds between the lattice constant and the long wave end of the absorption spectrum (1, p. 434) for the series of alkali halides sodium fluoride, potassium chloride, rubidium bromide*, which correspond to the metals sodium, potassium and rubidium with respect to closed electron shells. Indeed the actual wave-lengths at which absorption begins for the halides are not very different from those for the corresponding metals. It is also striking that for both series the onset of absorption is a gradual one. These facts suggest that the closed shell structures determine the optical properties for these wave-lengths. If this is the case the most recent form, advanced by Wilson (11), of the theory of the optical properties of metals will not apply to the alkali metals, which ought to offer the best chance for the successful application of a theory based on the free electron model. The first two absorption bands of the metal crystallizing in the body-centered cubic system, according to the theory, are given in Table II where the frequency ν is expressed in terms of $h/2ma^2$, where h = Planck's constant, m = mass of electron, a = lattice constant (side of

TABLE I

—	a (Å)	λ (Å)	λ/a
Cs	6.13	1860	303
K	5.32	1500	282
Na	4.28	1250	292

† *Note added in proof. In a letter to one of the writers Professor Wood has kindly stated that he has believed for some time that his supposed lithium films were really sodium.*

* *Lithium would have to be compared with lithium hydride and caesium with caesium iodide. The data for the former are not available and the latter is body-centered (type B_2), whereas the other halides are face-centered cubic (type B_1).*

elementary cube). The experimentally observed transmission extends from 0.73 to about 1.25 on the same scale, the latter figure varying with a. The first theoretical absorption band overlaps the transmission band actually observed, and the second absorption band is too far in the ultra-violet to explain the absorption observed to begin in the Schumann region. On the other hand, Kronig's theory (3) of the transmission (a modification of Zener's application (14) of classical optical theory) is supported by recent measurements of the optical properties of potassium in the near ultra-violet by Ives and Briggs (2)*. Kronig's theory provides the small absorption by the metal necessary for the photoelectric effect without introducing the lattice structure of the crystal, whereas Wilson's theory depends essentially on this structure. The critical wave-length for the beginning of transmission in the near ultra-violet is, according to Kronig and Zener, proportional to $a^{3/2}$, but agreement with experiment (a^2) can be achieved by a plausible adjustment (2). If this be accepted, the mechanism of Kronig appears to give a good account of the role of free electrons in determining the optical properties of the alkali metals in the near ultra-violet. It appears to the writers that in the far ultra-violet it is necessary to consider the metal on the same basis as insulating crystals, and to depend on "bound electrons" to explain the observed optical absorption.

TABLE II

Band number	ν min.	ν max.
1	0.24	3.76
2	1.52	6.50

Acknowledgment

The writers wish to thank Dr. A. N. Shaw for his kind interest in this work.

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* *The agreement with experiment claimed by Sergeiev and Tchernikovsky (5) cannot be accepted as significant.*

INTENSITY MEASUREMENTS IN THE DIFFRACTION OF LIGHT BY ULTRASONIC WAVES[1]

By F. H. Sanders[2]

Abstract

Measurements of the distribution of light energy among the various orders of the diffraction pattern produced when monochromatic light is passed through a liquid subjected to high-frequency ultrasonic disturbances have been carried out over a range of ultrasonic intensities at frequencies in the region of 5 megacycles per second. Both progressive and standing wave fields have been studied. Results of the experiments show excellent agreement with the theory of Raman and Nath for the variation in degree of scattering with varying ultrasonic intensity. Absolute measurements of the sound intensity conducted with a torsion pendulum are in good agreement with that expected from the theory for the liquids and light wave-lengths involved.

Introduction

The possibility of producing diffraction of light in a transparent medium subjected to periodic variations in density was predicted by L. Brillouin (9) in 1921. It was not until 1932 that an experimental demonstration of the phenomenon was realized by Debye and Sears (11) in the United States and by Lucas and Biquard (13) in France. The method pursued by both sets of investigators was the production of ultrasonic waves of very high frequency (10^6 to 10^7 cycles per second) in liquids by the use of the vibrations of a piezoelectric crystal. When a beam of parallel light was passed through the liquid in a direction normal to the direction of propagation of the ultrasonic disturbance, the light was scattered as by a plane diffraction grating. One of the interesting features observed was that, while Brillouin's theory had predicted the appearance of a single order of diffraction, the experiments showed a number of orders scattered according to the simple grating formula,

$$n\lambda = d \sin \theta$$

where d, the grating space, is equal to the wave-length of the ultrasonic disturbance in the liquid.

In an attempt to explain the divergence between theory and experiment a number of theoretical analyses dealing with the diffraction of light in a progressive wave field have been published. Debye (12) again obtained a result indicating the presence of a single order of diffraction. As an explanation of the multiple orders actually observed, Debye suggested that possibly a non-linear relation between density and dielectric constant might exist at high frequencies, or that the presence of harmonics in the vibration of the piezoelectric crystal might produce a number of single-order diffraction patterns of appropriate dispersion. Lucas and Biquard (14, 15) have pointed out that both proposals are unlikely, the first because of the relatively small pressure amplitudes involved, and the second because of the fact that a piezoelectric crystal will resonate only on odd harmonics.

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The latter writers have developed a theory (14, 15) based on a mirage effect, in which they obtain the amplitude and phase of an individual light ray as it progresses through the disturbed medium. This theory predicts multiple orders, the number of which increases with the length of path in the medium and the ultrasonic intensity. The distribution of light energy predicted by the above-mentioned theory, however, for path lengths and ultrasonic intensities where the light rays have not reached a focus in the sound field, is such that the relative intensities of the orders decrease monotonically with increasing order number. That this is not always the case has been shown experimentally by R. Bär (2). Under certain conditions of intensity, path length and optical wave-length, Bär has observed patterns in which some orders have far lower intensities than those of higher number.

Brillouin (10), enlarging on his earlier analysis, has also developed a theory giving multiple orders, but the mathematical difficulties restrict the application of the theory to the case where the intensity of the first order is still small compared with that of the central order.

In some very recent papers Raman and Nath (16, 17) have treated the scattering of light by a periodically disturbed medium in a manner analogous to that developed by Rayleigh for the diffraction of a plane wave incident normally on a periodically corrugated surface. The results of these theories are very interesting in that, in addition to predicting a dependence of the spectral character of the diffracted light on the number of the order both for progressive and standing waves, a type of distribution of the relative intensities of the various orders of light scattered by a standing wave field is obtained which differs very markedly from that to be expected with progressive waves. The scattering patterns for the progressive wave case appear to be in good qualitative agreement with those photographed by Bär.

H. Becker (5) has recently published a paper in which Debye's theory, with some modification, is compared with the results of an experimental study of the variation in intensity of the first order with ultrasonic intensity. For low sound intensities the agreement is quite good.

The present paper is a description of an investigation of the relative intensities of the various orders for ultrasonic intensities ranging from zero to values sufficiently high to give five or six orders of measurable intensity in both standing and progressive wave fields.

Experimental

Two different quartz crystals, one having a fundamental frequency of 370 kc. and the other of 448 kc., were used alternatively as a source of ultrasonic vibrations. The faces of the crystals were silvered chemically and copper-plated to ensure a continuous conducting surface. The crystal in use was supported vertically in the liquid by means of light phosphor-bronze contacts and excited by a "push-pull" oscillator employing two 200-watt Mullard transmitting tubes.

A Zeiss electric sodium vapor lamp operated on batteries provided a very steady and intense source of monochromatic light. A parallel beam of light

of dimensions 10 mm. by 10 mm. was passed through the sound field and focused upon a screen containing a narrow slit of such dimensions as to receive the light from a single order. Intensity measurements were effected by means of a photoelectric cell mounted behind the slit, the slit and photo-cell being movable transversely across the diffraction pattern. Since the light intensities frequently fell to very low values, a d-c. amplifier was used in conjunction with the photo-cell. The characteristic curve of the combination of cell and amplifier was obtained by the use of an inverse-square bench, and it proved to be quite linear over the range of light intensities concerned.

Several methods were used in the measurement of ultrasonic intensity. A high resistance in series with a vacuum thermocouple was connected across the crystal holder, and the square of the applied voltage read directly by means of a microammeter. Following some preliminary experiments that indicated the need of an intensity-measuring device that could be more readily adjusted to suit various intensity ranges, it was found more convenient to use a tuned circuit loosely coupled with the oscillator. When checked against each other these two methods showed excellent linear agreement.

As an additional check on the ultrasonic intensity measurements, a torsion pendulum was constucted which could be placed in the sound field just beyond the intersection of the light beam with the field. The pendulum vane was maintained in a vertical plane by a light but rigid shaft with bearings above and below, and was connected by a fine tungsten suspension to a calibrated torsion head. Since the torsion pendulum, having a measured reflection coefficient very close to 100% at the frequencies used, would set up standing waves when placed in the field, measurements with the pendulum were made only in the case of standing wave fields. The torsion pendulum was in all cases located very near the reflector producing the standing wave field; consequently the sound intensity in the field when the torsion pendulum vane was parallel to the crystal should not be appreciably different from that due to the reflector alone. This point was checked by measurement of the intensity of the central diffraction order with the reflector alone in place and with the pendulum vane in the parallel position. The photo-cell currents for the two cases checked to well within the errors of measurement. The force producing the deflections of the torsion pendulum should be due to the radiation pressure and hence directly proportional to the ultrasonic intensity. In the measurements, the torsion head was set at a definite angle and the output of the crystal adjusted until a balance was obtained. This eliminated any change in output due to the added capacity introduced by the presence of the operator's hand on the torsion head. Curves of torsion head settings were plotted against microammeter readings obtained by the two earlier methods and in all cases gave excellent straight lines. In addition to serving as a check on the linearity of the electrical measurements of relative ultrasonic intensity, the torsion pendulum provided a means of effecting an approximate determination of the absolute intensities involved.

Because of the difficulty of completely eliminating standing waves in a container of ordinary dimensions, the first portion of this investigation was

carried out with a standing wave field. The container used was a small biological cell of white mirror glass, 10 cm. in length. One end of the cell served as the reflecting surface and, by the use of a universal mount, could be set parallel to the face of the crystal. The crystal, also in a universal mount, could be adjusted quite independently of the reflector. In addition the whole system could be rotated as a unit about a vertical axis through the centre of the intersection of light beam and sound field. By means of a slow motion screw the reflector could be moved in the direction of propagation of the ultrasonic waves so that the optimum standing wave condition could be realized. Actually it was found that with these very short waves the resonance points were very broad and the change in degree of scattering as the reflector was moved quite small.

In order to obtain a sensibly pure progressive wave field a tank one metre in length was constructed. Taking Biquard's value of 0.047 for the absorption coefficient of xylol at 7.96 megacycles and reducing this to 4.94 megacycles, the lowest frequency employed, by assuming the absorption coefficient directly proportional to the square of the frequency, its value at this frequency would be about 0.018. Since the crystal was located within a few centimetres of one end of the tank, the intensity of the wave reflected from the far end of the tank would accordingly have fallen to something less than 10^{-3} of that originating at the crystal. The field through which the light passed could thus be regarded as consisting essentially of progressive waves. The same universal mount was used for the crystal as in the case of standing waves.

Measurements of optical intensity of both positive and negative orders from zero to five were carried out in xylol and petroleum ether as a function of ultrasonic intensity at frequencies of 4.06, 4.82 and 5.56 megacycles with standing waves, and at frequencies of 4.94, 5.84 and 6.72 megacycles with progressive waves. The above frequencies corresponded to the 11th, 13th and 15th harmonics of the two crystals. With the power available, ultrasonic intensities giving as high as 10 orders could be obtained, but as indications of cavitation and other disturbances in the liquid began to appear, the measurements were restricted to sound intensities giving rise to five or six orders of measurable intensity. Normally incident light was employed in all cases.

For each frequency a torsion pendulum curve was obtained at the end of the run, one of the electrical methods being used to measure the ultrasonic intensity during the course of the measurements of the diffraction patterns. The reflection coefficient and characteristics of the torsion pendulum were carefully determined so that a calculation of the absolute intensity in watts per cm.^{2} could be effected. In order to ascertain whether or not the standing wave field in the small tank suffered any distortion due to possible reflections from the side walls of the biological cell, the standing wave measurements were repeated in the large tank with a plane glass reflector placed at a distance from the crystal equal to that existing between the crystal and the end of the small tank. Results obtained at a frequency of 4.94 megacycles in the large tank checked those obtained in the small cell at 4.06, 4.82 and 5.56 megacycles to well within the errors of measurement.

Results and Discussion

The theory of Raman and Nath, referred to in the introduction, takes account of both progressive and standing wave fields. In the case of progressive waves the theory predicts the appearance of multiple orders, the frequency of any given order being changed by the Doppler effect of the moving sound waves from ν, the frequency of the incident light, to $\nu+n\nu^*$ for the positive orders and $\nu-n\nu^*$ for the negative orders, n being the number of the order and ν^* the frequency of the ultrasonic disturbance. The intensity of the mth order relative to that of the nth is given by the relation

$$\frac{I_m}{I_n} = \frac{J_m^2(v)}{J_n^2(v)} ,$$

where J_m and J_n are the Bessel's functions of order m and n, and the argument is given by

$$v = \frac{2\pi L \delta n}{\lambda} ,$$

where L is the length of path in the disturbed medium, δn the maximum variation in refractive index, and λ the wave-length of the incident light. From the nature of the Bessel's functions it is evident that, under certain conditions, some orders must fall to intensities much lower than those of orders of higher number.

In the case of standing waves, however, each order, instead of having a single frequency, consists of a series of sub-components of frequencies $\nu \pm 2r\nu^*$ for the even orders and $\nu \pm (2r+1)\nu^*$ for the odd orders, r being a positive integer. Thus, in the case of progressive waves, the light from any two orders should be completely incoherent, whereas, in the standing wave case, any two even orders should be partially coherent, a positive and negative order of a similar number should be completely coherent and any even order should be completely incoherent with any odd order. An experimental study of the coherence relations among the various orders produced in a standing wave field has been made by R. Bär (3). The results bear out the above-mentioned theory exceedingly well.

Very recently L. Ali (1) has made direct measurements of the shift in wave-length of the various orders due to diffraction by progressive waves, and has found quite good agreement with the theory for the latter case. The relative intensity of the $\nu \pm 2r\nu^*$ sub-component in the $2n$th order is given by $J^2_{n-r}(v/2)\, J^2_{n+r}(v/2)$ and that of the $\nu \pm (2r+1)\nu^*$ sub-component in the $(2n+1)$th order by $J^2_{n-r}(v/2)\, J^2_{n+r+1}(v/2)$. The total intensity of the mth order to that of the nth is given by the relation

$$\frac{I_m}{I_n} = \frac{\int_o^{2\pi} J_m^2 (v \sin\theta)\, d\theta}{\int_o^{2\pi} J_n^2 (v \sin\theta)\, d\theta} ,$$

and may be obtained by summation of the various sub-components in each order.

Variation with Relative Ultrasonic Intensity

In Fig. 1 the results of observations made with the photo-cell in a standing wave field at a frequency of 4.06 megacycles are shown. The liquid employed was petroleum ether. Each graph shows the diffraction pattern for a certain value of the ultrasonic intensity. Light intensities are expressed as actually observed in percentage of the intensity of the incident unscattered beam, rather than in percentage of the most intense order in any pattern as in the graphs shown by Raman and Nath. Since, for constant path length and light wave-length, the parameter v in the above-mentioned theory is directly proportional to δn, the maximum variation in index of refraction, and this is proportional to the square root of ultrasonic intensity, observations of light intensities were made as a function of the latter quantity. This arbitrary scale was reduced to the units of v by the choice of a suitable multiplying factor giving closest agreement between observed and theoretical values of light intensity over the whole range of ultrasonic intensities. The figures in the upper right-hand corner of each graph give the values of v thus obtained. The solid vertical lines represent intensities obtained from the Raman-Nath theory, while the circles represent observed data. The agreement is surprisingly good over the whole range. Careful examination of the theoretical intensities reveals the fact that the decrease in relative intensity with increasing order number is not quite monotonic for the higher

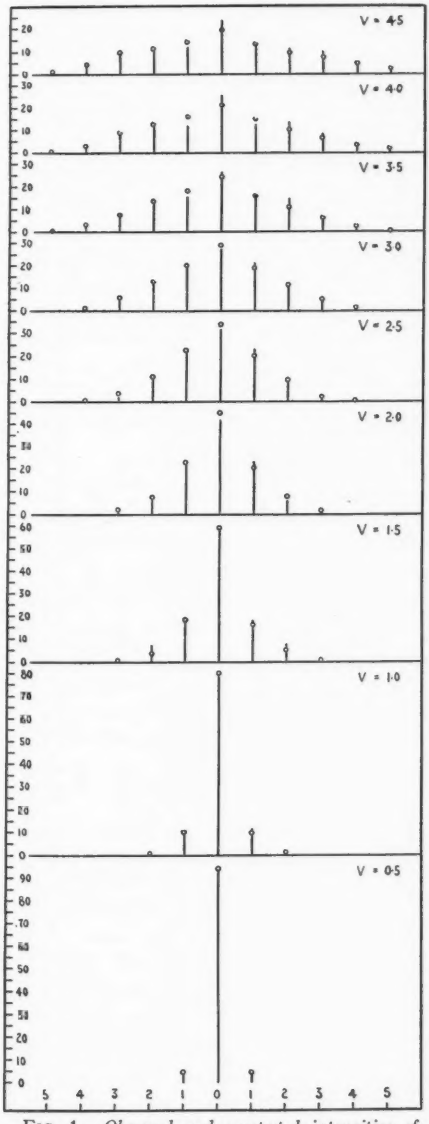


FIG. 1. *Observed and computed intensities of five orders obtained with standing waves. Each graph refers to a single value of the Raman-Nath v. Ordinates—per cent of incident light intensity. Abscissas—positive and negative order numbers.*

values of v, whereas the observed intensities do decrease monotonically. This deviation is undoubtedly due to the imperfections of the actual sound field. Since the ultrasonic beam is slightly divergent and a certain amount of attenuation in the liquid exists, the actual sound field is not quite the ideal rectangular prism of plane waves postulated in the theory. At the higher values of v a certain lack of symmetry in the intensities of positive and negative orders is noticeable. This assymetry persisted in all the measurements and appeared to be due to some type of distortion in the field. While it was possible to produce a very marked lack of symmetry by setting the crystal, reflector and light beam out of parallelism, under no conditions was it possible to obtain complete symmetry at all ultrasonic intensities.

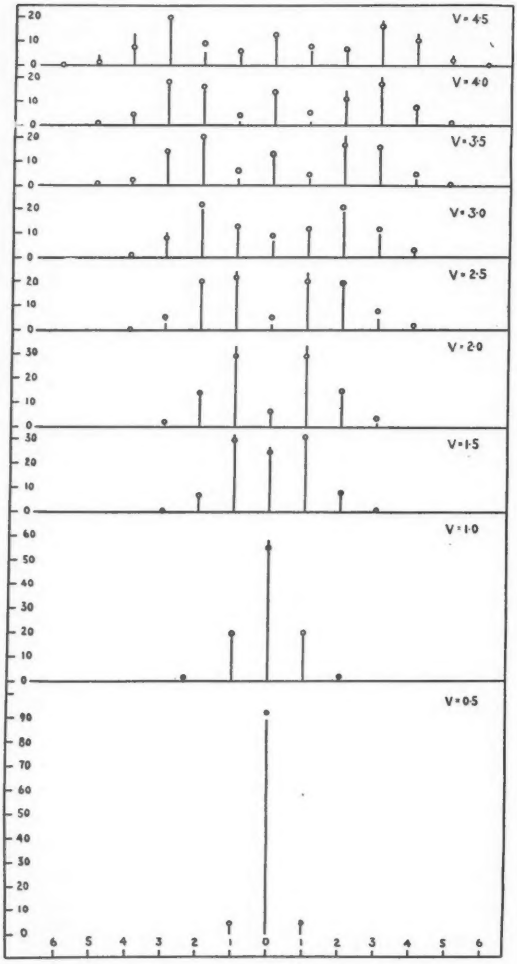


FIG. 2. *Observed and computed intensities of five orders obtained with progressive waves. Each graph refers to a single value of the Raman-Nath v.*
Ordinates—per cent of incident light intensity.
Abscissas—positive and negative order numbers.

Fig. 2 shows a similar set of data obtained with xylol, using progressive waves, at a frequency of 4.94 megacycles. As before, the solid lines represent intensities computed from the theory and the circles the observed values. The difference in nature of the patterns produced in a progressive wave field from those due to standing waves is quite striking. The central order falls to a very low value at $v=2.5$ and later increases to a second maximum. The first order goes through a similar cycle and the second has begun to decrease in intensity at the highest value of v. The choice of the multiplying factor converting square root of ultrasonic intensity to the units of v is considerably

simplified in the case of progressive waves, since it is necessary only to compare the observed ultrasonic intensities at which the central order attains its minimum and maximum values with the corresponding values of v obtained from the theory. A slight lack of symmetry is again observable at the higher values of v and could not apparently be eliminated.

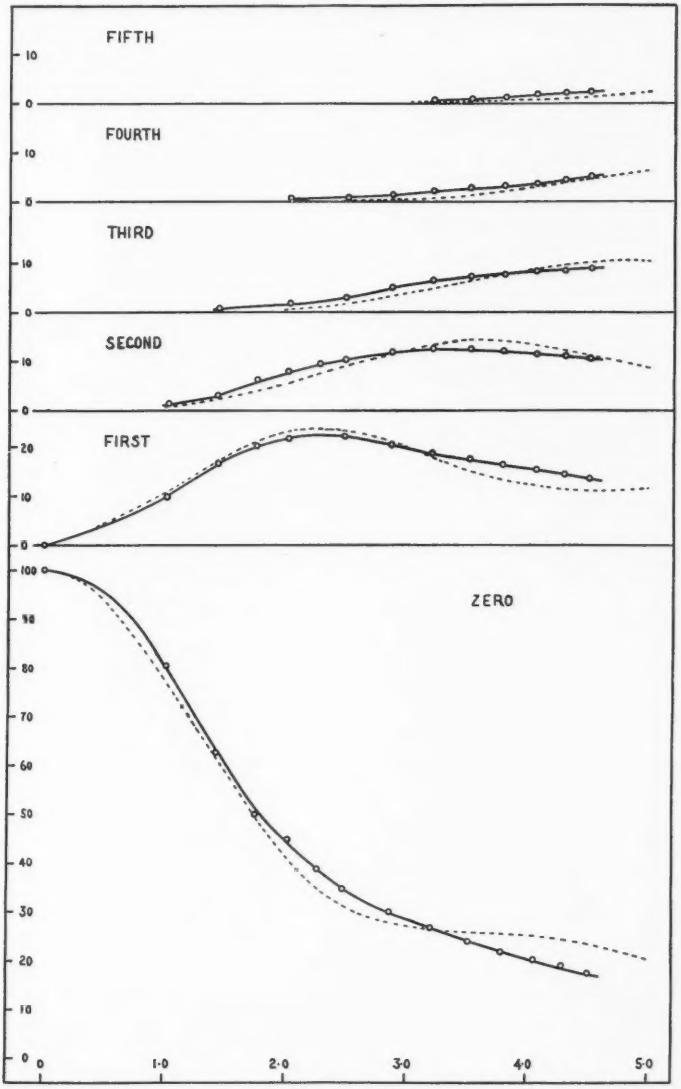


FIG. 3. *Continuous variation in light intensity with increasing values of the Raman-Nath v for a standing wave field. Each graph refers to a single order.*
Ordinates—per cent of incident light intensity.
Abscissas—Raman-Nath v values.

Figs. 3 and 4 show the progressive variation in intensity of the various orders with increasing values of v for standing and progressive waves respectively. The dotted lines represent theoretical values of light intensity, while the solid curves through the circles represent the mean value of each pair of positive and negative orders observed.

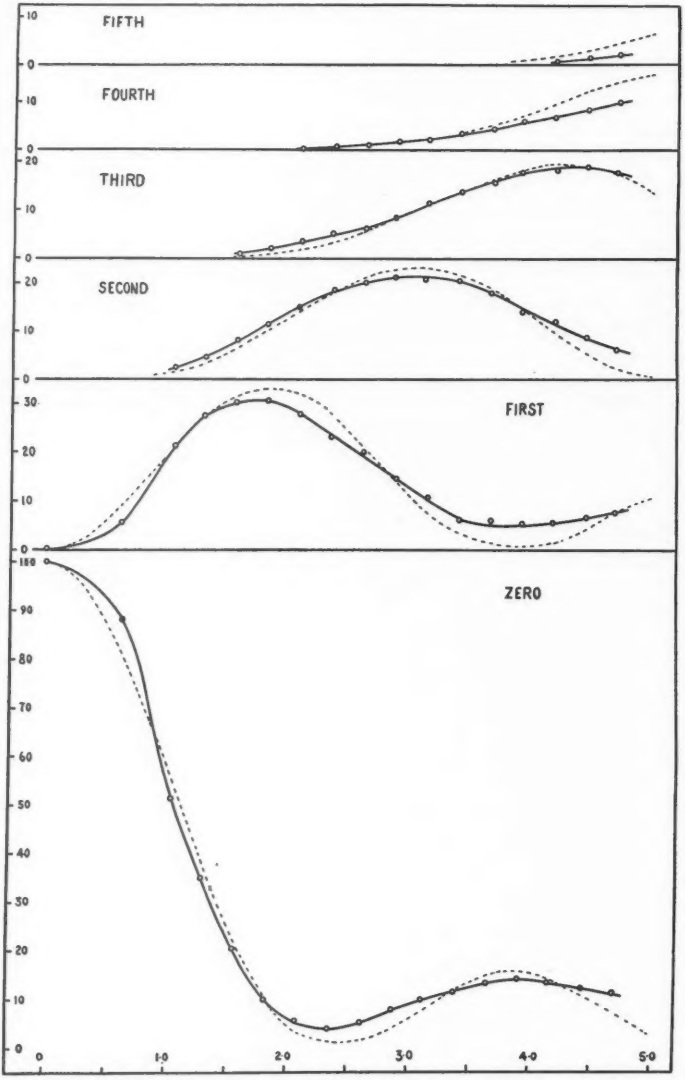
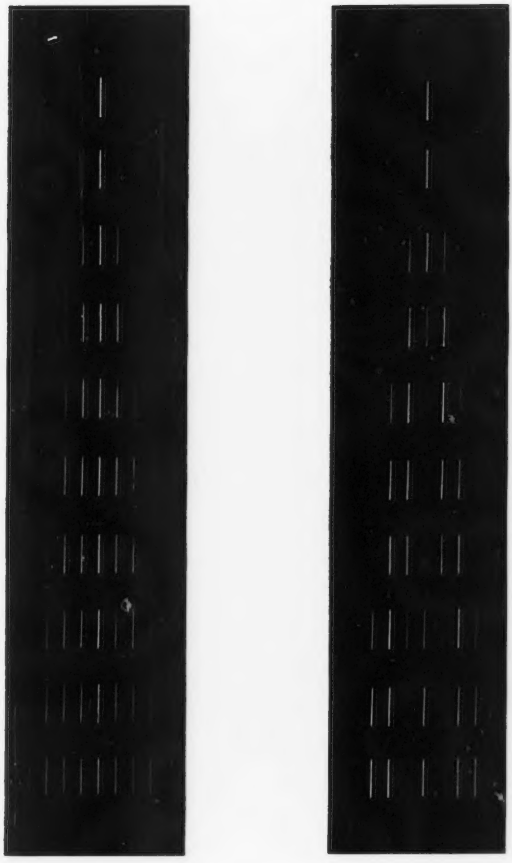


FIG. 4. *Continuous variation in light intensity with increasing values of the Raman-Nath v for a progressive wave field. Each graph refers to a single order.*
Ordinates—per cent of incident light intensity.
Abscissas—Raman-Nath v values.

PLATE I



a *b*

FIG. 5. *Photographs of diffraction patterns, (a) standing waves, (b) progressive waves. Exposures made at ten ultrasonic intensities corresponding to values of v ranging from 0 to approximately 4.0 in equal steps.*

The frequencies at which these observations were made are respectively the same as those of Figs. 1 and 2. The observed curves, because of the imperfections of the actual sound fields, have lower maxima and higher minima than those computed, but the general agreement is extremely good.

Similar curves were obtained at frequencies of 4.82, 4.94 and 5.56 megacycles with standing waves and at 5.84 and 6.72 megacycles with progressive waves. The agreement with the theory was equally satisfactory in all cases.

Fig. 5 shows two photographs, each consisting of 10 exposures, (*a*) of the standing and (*b*) of the progressive wave patterns. Exposure times were so estimated from the intensity values obtained with the photo-cell as to give approximately the same intensity to the most intense order in each pattern for all the exposures. The exposures were made for values of v from zero to about 4.0 advancing in steps of approximately 0.45. Comparison of the progressive wave patterns with the graphs shown by Raman and Nath for the above values of v shows a very satisfactory qualitative agreement.

Absolute Measurements

From the constants and deflections of the torsion pendulum, some approximate determinations of the absolute energies involved were made for the standing wave field in petroleum ether. The velocity of sound in this liquid, its density and index of refraction, were determined experimentally. With this information it was possible to determine the energy density in watts per cm.^{2} and consequently the maximum variation in index of refraction. Knowing the length of path in the disturbed medium and the wave-length of the light used, it was possible to evaluate the quantity v. The quantities used were as follows: velocity, 1.15×10^5 cm./sec.; density, 0.633; index of refraction, 1.365; length of path, 2.3 cm.; light wave-length, 5.89×10^{-5} cm. Table I shows the nature of the agreement between the computed values of v and those for which the observed light intensities gave closest agreement with the theory.

TABLE I

Frequency, megacycles	Energy density, watts/cm.^{2}	Maximum variation in refractive index	v, from torsion pendulum data	v, observed
4.06	0.171	1.34×10^{-5}	3.29	4.50
4.82	0.106	1.05×10^{-5}	2.58	4.00
5.56	0.183	1.38×10^{-5}	3.40	4.20

In view of the inaccuracies of absolute measurements at these frequencies the agreement is quite satisfactory.

Number of Orders Visible as a Measure of Ultrasonic Intensity

While investigating the effect of ultrasonic intensity on the degree of diffraction, it appeared of interest to determine the manner in which the number of orders visible in the diffraction pattern of a standing wave field varies with ultrasonic intensity. R. Wyss (18) has made use of the number

of orders visible when a light beam is passed through a sound field at varying distances from the source, to measure the relative absorbing powers of various liquids. By means of an arbitrary scale of visually estimated intensities Wyss was able to obtain quite smooth curves showing the relative absorbing powers of ethyl alcohol, amyl alcohol, benzene, xylol and carbon disulphide.

Since it is difficult to define a threshold of visibility, certain arbitrary levels of light intensity were chosen in the writer's experiments as indicated by the deflections of the galvanometer measuring the intensities of the various orders. These levels corresponded to intensities of 0.57 and 0.86% of that of the unscattered beam. Fig. 6 shows two curves giving the number of

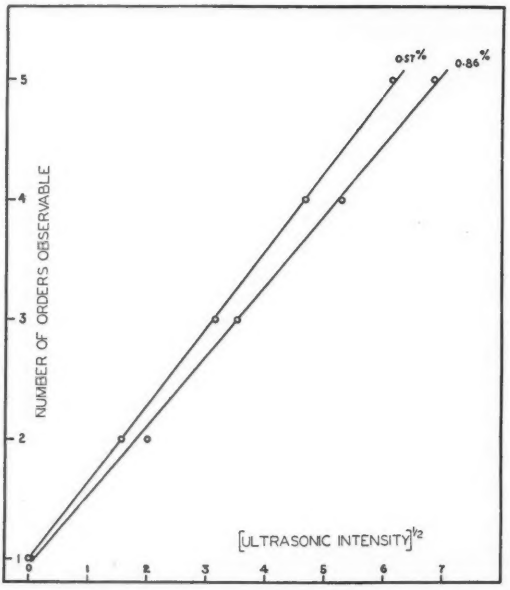


FIG. 6. *Number of orders having intensities equal to or greater than 0.57% and 0.86% respectively of the intensity of the incident light, as a function of ultrasonic amplitude.*

orders having an intensity equal to or greater than these arbitrary levels as a function of the square root of ultrasonic intensity. Each point was obtained by setting the photo-cell on the given order and determining the ultrasonic intensity for which the galvanometer gave the required deflection. It is evident that the number of orders observable, at least for orders up to five, is very closely a linear function of the square root of ultrasonic intensity and hence of ultrasonic amplitude. Examination of Raman and Nath's theory indicates that this relation is to be expected.

Knowing the above relation it is possible to evaluate the absorption coefficients of the liquids studied by Wyss. The natural logarithm of the ultrasonic amplitude plotted as a function of distance in centimetres should

give a straight line of slope equal to the absorption coefficient. Actually the only liquid for which Wyss' results give a good straight line is benzene; the two alcohols and xylol give curves having a slight upward concavity while the carbon disulphide curve shows a slight downward concavity. Taking the slope of the best straight line through each of the above curves, the values in Table II were obtained for the respective absorption coefficients at the frequency of 6.0 megacycles used by Wyss.

TABLE II

Liquid	Ethyl alcohol	Amyl alcohol	Xylol	Benzene	Carbon disulphide
Absorption coefficient	0.057	0.069	0.071	0.33	4.3

Of the liquids listed in Table II, benzene and xylol have been studied by Biquard (6, 7) who used a frequency of 7.96 megacycles. If Biquard's values are converted from 7.96 to 6.00 megacycles, and it is assumed that the absorption coefficient is proportional to the square of the frequency, the values obtained are 0.027 for xylol and 0.33 for benzene. The very close agreement between the two values for benzene is undoubtedly fortuitous, but the order of magnitude in both liquids is correct and serves to corroborate the abnormally high values of absorption coefficient reported at these frequencies. R. Bär (2) has already drawn attention to the excessively high absorption in carbon disulphide. A more complete study of absorption coefficients at various frequencies has been made by Biquard (8), but the writer has not yet been able to obtain a copy of the work.

Effect of Light Wave-length

According to the theory of Raman and Nath, the quantity v determining the degree of scattering is inversely proportional to light wave-length. Hence the effect of increasing the light wave-length should be equivalent to that obtained by decreasing the square root of ultrasonic intensity. A few measurements were conducted in a standing wave field with light of the wave-lengths 436, 546 and 589 millimicrons. An appreciable flattening of the diffraction patterns with decreasing wave-length was noted, but the range studied was too limited to give good quantitative agreement with the theory. It is hoped to conduct some further studies of this phase of the problem with both standing and progressive wave fields over a considerably larger range of wave-lengths.

Conclusion

From the results of the foregoing experiments it is evident that the theory proposed by Raman and Nath gives the correct type of envelope for the diffraction pattern produced by both standing and progressive wave fields in the frequency region investigated. The results of Bär and Ali regarding the spectral characteristics of the diffracted light appear further to substantiate this theory.

Bär (4) has, however, recently reported on some apparent discrepancies observed in the region of 7.5 megacycles. While the nature of the intensity distribution in the various orders observed by Bär appears to be in good qualitative agreement with the Raman-Nath theory, experiments carried out with the incident light inclined slightly to the planes of the sound waves give results that are not in accord with the above theory. In addition, Bär was able to obtain "photographs" of the actual ultrasonic grating by inserting a photographic plate or strip of sensitized paper in an appropriate position in the light beam just behind the stationary sound field, no lens system of any type being required to focus the light on the plate. From these observations it is apparent that the light rays in passing through the sound field undergo not only changes in phase, as assumed by Raman and Nath, but also some changes in amplitude as postulated by Lucas and Biquard. Bär notes that these discrepancies are considerably more apparent at a frequency of 7.5 megacycles than at 1.5 megacycles. Hence he concludes that Raman and Nath's assumption of phase changes alone is not justified, particularly at the higher frequency.

Since the writer's quantitative measurements demonstrate the validity of the Raman-Nath intensity distribution for frequencies ranging from 4 to 7 megacycles, there appears at first to be a contradiction in the experimental results. It seems possible, however, that these apparent anomalies may be explained as follows:

In the theory proposed by Raman and Nath, the type of sound field postulated is a rectangular prism of plane waves progressing in a direction normal to that of the incident light, the amplitude of the variation in refractive index at any instant being constant across the field in the direction of motion of the light rays. A single light ray is hence incident normally on a medium of constant refractive index. Under such a condition the light can suffer changes in phase only, and the Raman-Nath assumption is quite valid. The sound field produced by an oscillating crystal, however, is not quite of this ideal form. It is well known that the intensity distribution across the ultrasonic beam is by no means constant, while there is certainly not a plane surface of discontinuity at the edge of the field. Consequently a light ray, even though incident in a direction normal to the axis of the ultrasonic beam, will suffer some refraction on entering the sound field. Once bent, the ray will be passing through layers of varying refractive index and will suffer changes in amplitude as well as in phase. If such is the case, the rays may eventually come to a focus, producing the fringes observed by Bär and others. Furthermore, the Raman-Nath theory for oblique incidence, which is dependent on the existence of a rectangular sound field, will not be valid. In the case of normally incident light, however, it is quite conceivable that small changes in amplitude, combined with changes in phase of the light rays progressing through the liquid, will give rise to an emergent light wave surface that approximates very closely to that which would be produced if changes in phase alone occurred. That such is the case seems to be borne out by the

close agreement between the observed envelopes of the diffraction patterns and those predicted by the Raman-Nath theory. Bär's observations of the fact that the discrepancies noted at 1.5 megacycles are much smaller than those at 7.5 megacycles seem to lend support to the above hypothesis, since the longer the sound wave-length the higher the probability of the light entering under the conditions postulated by the theory.

It would thus appear that while Raman and Nath's fundamental assumption is not entirely true for a practical sound field, the nature of the emergent light wave surface is very much that which would obtain if changes in phase alone occurred, so that the intensity distribution in the various orders of the diffraction spectrum is very closely that predicted by the above theory. From the results of Bär and the writer, it would appear that this is true for frequencies ranging from 1.5 to 7.5 megacycles. Until a theory can be developed which will take complete account of the true nature of a practical sound field, it would appear that the theory of Raman and Nath approximates most closely to the observed diffraction effects.

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